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Mobile platform measurements of ultrafine particles and associated pollutant concentrations on freeways and residential streets in Los Angeles

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Abstract

Recent health studies have reported that ultrafine particles (UFP) (<0.1 μm in diameter) may be responsible for some of the adverse health effects broadly attributed to particulate matter. In urban areas, UFP are produced by combustion sources, such as vehicle exhaust, and by secondary formation in the atmosphere. While UFP can be monitored, few studies have explored the impact of local primary sources in urban areas (including mobile sources on freeways) on the temporal and spatial distribution of UFP. This paper describes the integration of multiple monitoring technologies on a mobile platform designed to characterize UFP and associated pollutants, and the application of this platform in a study of UFP number concentrations and size distributions in Los Angeles. Monitoring technologies included two condensation particle counters (TSI Model 3007 and TSI 3022A) and scanning mobility particle sizers for UFP. Real-time measurements made of NO_x (by chemiluminesence), black carbon (BC) (by light absorption), particulate matter-phase PAH (by UV ionization), and particle length (by diffusional charging) showed high correlations with UFP numbers, ($r^2 = 0.78$ for NO, 0.76 for BC, 0.69 for PAH, and 0.88 for particle length). Average concentrations of UFP and related pollutants varied strongly by location, road type, and truck traffic volumes, suggesting a relationship between these concentrations and truck traffic density.

Keywords: Particle number concentrations; On-road concentrations; In-vehicle concentrations; Nanoparticles

1. Introduction

Airborne particles, most often measured as the mass of particles <10 or 2.5 μm in diameter (PM10 and PM2.5), are associated in epidemiological investigations with a range of adverse health outcomes including increased illness, hospitalizations, and mortality rates (e.g., Dockery et al., 1993; Burnett et al., 2000; Samet et al., 2000; Dockery, 2001; Pope et al., 1995, 2002; Schwartz et al., 2002; Brunekreef and Holgate, 2002; de Hartog et al., 2003). Recent health studies have reported that ultrafine particles (UFP), with diameters <0.1 μm, may be responsible for some of these health outcomes, even though they account for a very small portion of total PM mass (Donaldson et al., 1998; Obersdörster

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et al., 2000; Obersdörster, 2001; Penttinen et al., 2001). UFP are composed of many constituents, including metals, organic compounds, carbon, and acid aerosol, some of which are toxic. UFP also have a high surface-to-volume ratio, are not efficiently removed by macrophage clearance mechanisms in the lung, and may cause mechanical damage to lung tissue (Donaldson et al., 1998; Hughes et al., 1998; Utell and Framptom, 2000; Seaton and Dennekamp, 2003). Experimental exposure studies have demonstrated that inhaled UFP are deposited much more efficiently in the lungs than larger particles such as PM10 or PM2.5 (Brown et al., 2002; Jaques and Kim, 2000; Kim and Jaques, 2000; Smith et al., 2001) and can be transported through the blood stream to the heart, brain, and other organs (Nemmar et al., 2002; Obersdörster et al., 2002).

Two sources appear to dominate UFP levels in ambient urban air: combustion emissions from sources such as vehicular traffic or power plants, and atmospheric chemical reactions of gaseous pollutants such as SO_2, NO_x, ammonia and organic vapors. Studies have demonstrated pronounced UFP gradients near sources, such as the rapid decrease in particle counts with distance downwind of freeways (Zhu et al., 2002a, b). Particle counts appear to reach background levels in as little as 300 m distance downwind. Because of sharp gradients, sparsely distributed, fixed-site monitoring does not provide a means to establish the spatial nature of UFP.

Recent studies have developed mobile monitoring platforms using real-time instrument technologies to assess on-road air quality as well as the spatial and temporal gradients of air quality in urban areas. Mobile monitoring studies that included UFP measurements have been performed in and near Zurich, Switzerland (Bukowiecki et al., 2002a, b, 2003); throughout the Netherlands (Weijers et al., 2004); and in Helsinki, Finland (Pirjola et al., 2004). In the United States, such studies have only been performed in New York City (Canagaratna et al., 2004), Minneapolis (Kittelson et al., 2004a) and a Rochester-to-Buffalo interstate loop in New York (Kittelson et al., 2004b). The studies of Canagaratna et al. (2004) and Kittelson et al. (2004b) purposefully followed or "chased" specific vehicles, while the study by Kittelson et al. (2004a) purposefully avoided vehicle exhaust plumes. The purpose of the current research was to test a non-polluting, mobile-platform approach to characterizing UFP concentrations and associated co-pollutants during realistic (non-chase) driving in Los Angeles, the city with the heaviest traffic density in the United States. Compared to the mobile platform measurement studies listed above, it was unique in that it was the only United States study to measure roadway concentrations during realistic driving; it covered the widest range of traffic and truck traffic densities (e.g., 0.6 to >18% diesel trucks on freeways with >200,000 vehicles per day); and it used a non-polluting vehicle. It also utilized a relatively wide variety of instruments including the first use of two scanning mobility particle sizers (SMPSs) side-by-side, which allowed simultaneous measurements of the full size range of on-road PM2.5.

2. Methods

2.1. Mobile sampling platform

We used a 1998 electric Toyota RAV4 SUV as our mobile sampling platform because it was a zero emission vehicle, capable of transporting substantial weight, and easily modified to accommodate our instrument package. Air was sampled from an inlet, consisting of 6-in diameter galvanized steel duct, located 1.5 m above the roadway in the rear passenger space of the vehicle. Air was pulled into the duct by an inline fan located downstream of all sampling ports. Air entered the ducting through the rear window on the passenger-side, traveled through the manifold, passed the inline fan, and exited through the driver's side passenger window. Each instrument sampled from ports in the ducting, connected by conductive tubing. Sampling lines were kept very short (0.5–2 m) to minimize residence time and particle losses.

Instruments were powered by a 2-kW/115-V inverter connected to sealed lead-acid batteries. This power system provided up to 6 h of continuous instrument operation. The vehicle had a range of approximately 120 k at freeway speeds.

2.2. Monitoring equipment

Table 1 presents a list of instruments, parameters measured, and data averaging periods. Instruments placed in the RAV4 were selected for their high time resolution, their low power consumption, and when possible, their small size.

UFP number concentrations were measured with the TSI Portable Condensation Particle Counter Model 3007 (CPC), reporting particle counts in the size range of 0.01–1 μm. Because this unit did not have correction algorithms for particle coincidence at high concentrations (e.g., greater than 100,000 cm^{-3}) roadway-specific correction factors were developed by comparing simultaneous readings from a TSI model 3022A CPC, counting particles 0.007 μm and larger at concentrations up to 10^7 particles cm^{-3}. The 3022A was not used as the primary CPC because of its sensitivity to tilt or motion. Related to particle number concentrations, "total aerosol length" (or particle number times average diameter, reported as mm cm^{-3}) was measured with the TSI Electrical Aerosol Detector Model 3070a (EAD), which charged particles via unipolar diffusional

Table 1
Monitoring instruments employed in the mobile monitoring platform

Instrument	Parameter measured	Time resolution (s)
TSI portable CPC, model 3007	UFP count, 10 nm–1 μm	10
TSI CPC, model 3022A	UFP count, 7 nm–1 μm	10
TSI Electrical Aerosol Detector, model 3070A	Particle length mm cm^{-3}	2
Magee Scientific portable aethalometer, model 42	Black carbon	60
TSI scanning mobility particle system model 3080 classifier:	Particle counts size^{-1}	60 s per scan
Nano DMA, 3025 CPC	5–153 nm	
Long DMA, 3025 CPC	16–600 nm	
EcoChem PAH analyzer, model PAS 2000	Particulate matter-phase PAH (ng m^{-3})	2
TSI DustTrak	PM2.5	10
Teledyne-API NO_x analyzer, model 200e	NO, NO_x, NO_2	20
TSI Q-Trak Plus monitor, model 8554	CO, CO_2, Temp., humidity	10[a]

[a]Actual instrumental response: 60 s for CO, 20 s for CO_2.

charging and calculated length from the total charge collected.

UFP size distribution data were measured by one or more TSI SMPS systems, using long and/or short differential mobility analyzers (DMAs) to classify the particles by size, and a TSI model 3025 CPC, sensitive down to 0.003 μm. Scans were set for 60 s with a 15-s refresh period.

Other pollutants measured included PM2.5, black carbon (BC), particulate matter-phase PAHs (PM-PAHs), NO_x, CO and CO_2. Total PM2.5 mass was measured with a TSI DustTrak nephelometer with a PM2.5 cyclone inlet. BC was measured with a Magee Scientific Portable Aethelometer Model 42, operating with quartz filter tape, illuminated by 880 nm wavelength light, with the allowable optical attenuation depth of the filter set to 1.25, and a flow rate of 2.1 lpm. Oxides of nitrogen were measured with an API-Teledyne Model 200e analyzer, utilizing chemiluminesence and configured by the manufacturer to report data with 20-s time resolution. Carbon monoxide and Carbon dioxide were measured with TSI Q-Trak Model 8554 monitors, which also reported relative humidity and temperature.

PM-PAHs were measured with an EcoChem PAS 2000, detecting particle-bound PAHs with three or more rings through the measurement of electrons emitted by organic molecules on particles irradiated by UV light. Elemental carbon (EC) also contributes emissions of photoelectrons under UV irradiation, although to a lesser extent than PM-PAH. Therefore, for diesel particulate matter, with its high fraction of EC, the EcoChem PAS 2000 measurements should be considered PM-PAH with potentially significant contributions from EC (Burtscher, 2004).

2.3. Data display and storage

Concentration data for BC, NO_x (NO, NO_2, and total NO_x), PM-PAH, and aerosol length were stored and displayed on a Eurotherm Chessel Model 4100G video data-logger, and, where available, in an instrument's internal memory as back-up. UFP number count concentrations and size distributions were collected and stored on a PC, and CO and CO_2 data were stored in the Q-Trak's memory. When both analog and serial outputs were available, data outputs were also stored and displayed on a personal computer, to provide further back-up and to provide real-time display during study runs.

Videotapes of operations were made with a Sony Mini DV camera with a view through the front window. Tapes from this camera served as documentation of the road and traffic conditions, helped confirm and identify emission sources, and served as an oral record of driver observations.

2.4. Flow checks, time synchronization, and calibrations

Quality assurance measures included flow and zero checks of all instruments, and regular calibration of gaseous pollutant monitoring instruments. Flow measurements were made several times each week with an NIST traceable rotometer; in the final month of the study a lower-resistance and more accurate DryCal flow meter was used. Instruments and data logging devices were synchronized to a satellite-signaled clock. Time synchronization was adjusted as necessary by linking the satellite time as recorded on video with traffic events such as following high-emitting vehicles. The NO_x analyzer was calibrated with zero and span gas (750 or 800 ppb) twice weekly. The Q-Traks were calibrated at the start of the study with a series of 4 concentrations of CO generated from NIST traceable standards. These devices were also checked while in operation with vendor-supplied zero and span gases.

2.5. Driving routes and protocol

The RAV4 mobile monitoring platform was driven on fixed routes in the Los Angeles roadway network. The

primary route is shown in Fig. 1, with the 710 Freeway selected for its very high concentration of heavy-duty diesel trucks and the 110 (north of downtown) on which heavy-duty truck traffic is prohibited. The 10 and 5 Freeways had a mixture of light and heavy-duty traffic. At the ends of the freeway route in Long Beach and Pasadena data were also collected while parked in residential locations away from freeways or arterial roads. The freeway loop typically took about 2 h to complete and covered approximately 120 km. Normal driving was emphasized with no targeting of specific vehicles. The right-hand lane (slow or merge lane) was avoided similar to typical driving, i.e., used primarily for entrances and exits. Location was recorded with global positioning satellite system (GPS) every min.

3. Results and discussion

Measurements from 5 days are summarized in this article: data from 24 April 2003 demonstrate the performance of the Model 3007 CPC compared to the Model 3022A CPC, while data from 14 and 20 February and 7 and 16 April 2003 illustrate typical freeway results. Traffic and weather conditions on these 5 days were typical of springtime weekdays in Los Angeles, i.e., clear and dry with daytime temperatures near 21 C (70 F), with moderate on-shore breezes from the south and southwest.

Table 2 presents the wind speed and direction from the nearest of four meteorological stations (Pasadena, downtown LA, Long Beach, and Lynwood Station, about halfway between Long Beach and downtown LA along the 710 Freeway) for the hours in which a study vehicle passed the station or stations. Compared to the median wind speed and direction for the last 2 weeks in February, 2003 and the first 3 weeks in April 2003, the wind speeds during the study runs were typical except for April 7, where speeds were about half of typical. It is interesting to note that on this day, on-road measurements of CO and NO (columns 5 and 6) were much higher relative to other days while the nearest ambient networks (same locations and hours) showed much smaller differences. It is also interesting to note that roadway concentrations of CO and NO_2 were usually no more than about twice the ambient concentrations, while NO concentrations on roadways were 10–40 times higher.

3.1. 3007 CPC versus 3022A CPC

Our results demonstrated the hand-held 3007 CPC provided a simple, real-time UFP measurement that was well correlated with more traditional measurement

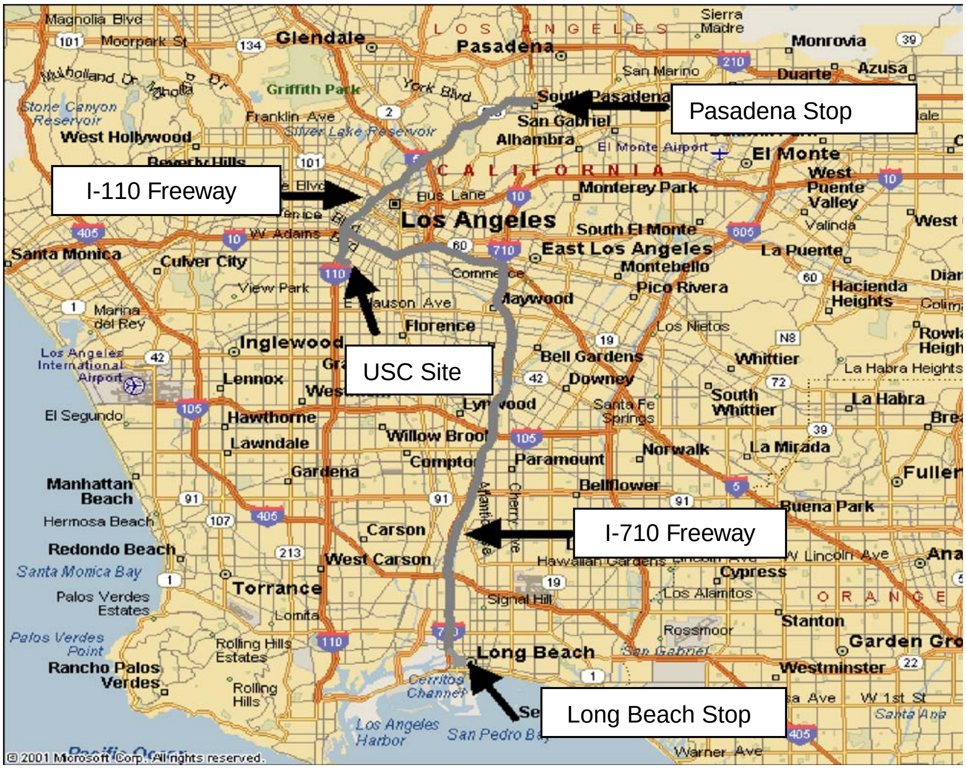

Fig. 1. Map of Los Angeles study area. Gray line represents freeway-sampling route.

Table 2
Wind speed and pollutant concentrations during runs compared to nearest meteorological or ambient monitoring stations

	Wind speed				Pollutant concentrations						Run time hours used
	During run		Nearest of four stations, all month[a]		On-road			Ambient monitors[a]			
	Speed ($m\,s^{-1}$)	Direction (deg)	Speed ($m\,s^{-1}$)	Direction (deg)	CO (ppm)	NO (ppb)	NO_2 (ppb)	CO (ppm)	NO (ppb)	NO_2 (ppb)	
14 February											
25th %	1.5	175	1.3	172	0.7	40	18	0.6	4.3	31	2–6 pm
Median	2.2	191	2.2	217	1.9	120	35	1.0	15	34	
75th%	2.6	216	3.1	239	2.9	240	62	1.1	19	35	
20 February											
25th%	2.2	169	1.2	138	0.2	60	11	0.3	2.0	7	11 am–1 pm
Median	2.7	235	1.8	171	0.8	160	31	0.6	4.0	12	
75th%	2.9	237	2.2	235	1.6	300	59	0.7	18	23	
7 April											
25th%	0.4	170	0.9	147	3.4	290	48	0.8	20	37	9 am–12pm
Median	0.9	210	1.8	186	4.1	400	55	0.8	22	39	
75th%	1.3	234	2.7	236	4.9	470	60	0.8	31	40	
16 April											
25th%	2.2	213	1.8	171	0.7	50	33	0.7	11	30	11 am–2 pm
Median	2.7	234	2.7	214	1.7	180	53	0.9	18	35	
75th%	3.6	238	3.6	238	2.4	370	82	1.0	32	40	

[a]From nearest meteorological or monitoring station (of four) for each hour of run.

techniques, although above 100,000 particles cm^{-3} the instruments differed in response. This was most likely due to the 3022A having a photometric mode that compensates for particle coincidence effects, while the 3007 has no such compensation. The scatter plot in Fig. 2 shows the non-linear relationship between the two instruments at high UPF concentrations and the best-fit logarithmic curve used to correct 3007 UFP concentrations for coincidence. The equation used for correction was

$$y = 38456e^{0.00001x} \text{ for } x > 100,000\,cm^{-3} \quad (r^2 = 0.817),$$

where x is the 3007 UFP count and y is the predicted 3022A UFP count. This correction factor was used in all 3007 UFP concentrations presented in the rest of this article.

Below 100,000 particles cm^{-3}, the 3007 slightly undercounted UFP compared to the 3022A, at least partly due to its size detection limit of 0.01 versus 0.007 μm for the 3022A. However, no adjustments were made to the 3007 CPC data below 100,000 as these differences may reflect real variability in particle numbers due to the different lower size limits of the instruments.

3.2. Pollutant concentration differences by location

Pollutant concentrations varied widely by location and/or roadway and appeared to be strongly affected by traffic sources. Table 3 presents average median and interquartile range concentrations (IQR, 25th and 75th percentile) for 4 days (2/14, 2/20, 4/7, 4/16) for several pollutants. The median and IQRs were used as they are less sensitive to extreme values or serial (temporal) correlation.

The Long Beach and Pasadena residential street locations where the vehicle was parked for short times had the lowest concentrations. The USC location, as the starting and finishing destination, had concentrations intermediate between residential areas and freeways or arterial roads. Although the USC location was away from traffic, it was in an area of high traffic density with a freeway about 120 m to the west and a busy arterial road about 100 m to the north. Most of the concentrations at USC were lower at the finish of runs, 2–4 h later, than at the start, and concentrations for NO, CO, and, CO_2 were significantly lower at USC after the run than for the arterial roads just north of USC driven near the end of the runs.

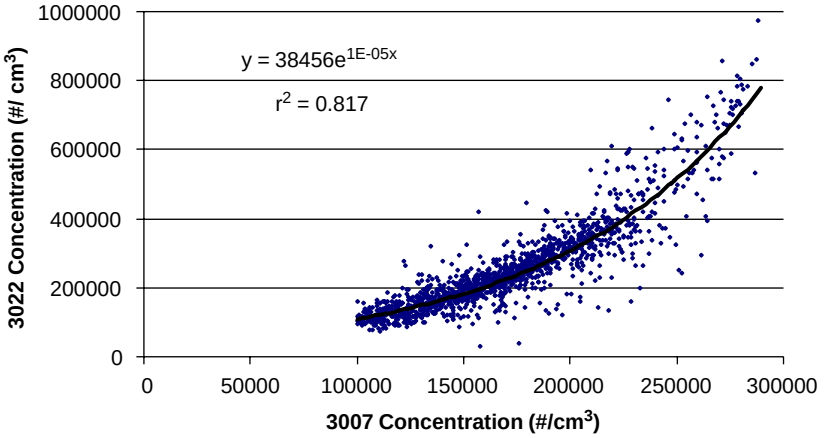

Fig. 2. Comparison of Model 3007 and Model 3022A data.

In contrast with other locations, the three freeway segments showed much higher concentrations: UFP, NO, BC concentrations were up to 20 times those measured in residential location, including CO_2 if background CO_2 concentrations (~370 ppm) are subtracted from the readings. UFP, NO, and BC concentrations also appeared to increase sharply as diesel truck traffic increased. The average diesel truck counts of 3500, 10,000, and 25,000 listed for the 110N, 10E, 710S, respectively, corresponded to 1.4%, 3.6%, and 14% of the total average daily traffic volumes (209, 273, and 182 thousand vehicles per day) on these roadways (CalTrans, 2004).

In contrast to UFP concentrations, PM2.5 showed much less variation in concentration by location or roadway, and was more influenced by specific encounters with high-emitting vehicles. While the medians reported in Table 3 are generally not affected by the occasional extreme concentrations produced by following high emitters, the influence of one particular high-emitter significantly affected the average median on the 10E Freeway. This occurred because the 10E Freeway segment was relatively short, PM2.5 was only measured on 2 days (14 and 20 February) and on 20 February, the segment was dominated by a high-emitting diesel vehicle that produced PM2.5 concentrations above $6000\,\mu g\,m^{-3}$.

In contrast to UFP, NO, and BC, both CO and CO_2 appeared to reflect proximity to all vehicles rather than just diesel vehicles, with the arterial road CO and CO_2 concentrations being similar to those of freeways and much higher than residential areas.

The averages in Table 2 were generally statistically significantly different between freeway and non-freeway locations, with the exception of BC and CO. For BC the most significant difference statistically was between roadways with significant diesel truck volumes (i.e., the 10E and the 710S) and other roadway and non-roadway locations (Tukey HSD test, $p = 0.05$). For CO, the most significant difference was roadways (freeways and arterial roads) versus non-traffic locations (i.e., residential and USC locations) (Tukey HSD test, $p = 0.05$). It is interesting that although Long Beach and Pasadena are about 42 km apart, they appeared to have rather similar average median concentrations for the pollutants listed.

3.3. Pollutant concentration correlations

Fig. 3 presents a scatter plot matrix of the major pollutants measured for the entire run on 16 April, including all freeway, arterial road, and residential road concentrations. UFP, particle length (EAD), BC, NO, and PM-PAH 10-s averages all showed high correlation with each other, while CO_2 showed moderate correlation and CO showed poor correlation.

The corresponding Spearman's ρ^2 and Pearson's r^2 values are shown in Table 4. Both the non-parametric Spearman's ρ^2 and the parametric Pearson's r^2 are presented in Table 4 for comparison but the Spearman values are the more appropriate single measure of correlation overall due to the presence of a extreme values for some of the pollutants due to concentration spikes from individual vehicle plumes. The ρ^2's were typically above 0.9 for the measurements most affected by diesel vehicle volumes (UFP, NO, BC, EAD particle length, and PM-PAH. The ρ^2's between CO_2 and the other pollutants were moderately good with ρ^2's mostly in the range of 0.6. Because CO_2 is emitted in high quantities from all fossil-fuel-powered vehicles, moderate correlation is not surprising. Likewise, the poor ρ^2's between CO and other pollutants, in the 0.2–0.3 range, reflect how CO emissions are primarily from gasoline-powered vehicles, and relatively unrelated to those pollutants dominated by diesel vehicles. Similar low

Table 3
Average median and average interquartile range (P_{25}, P_{75}) of pollutant concentrations by road segment or location for 4 days in Spring, 2003

Location	UFP (coincidence corrected) (1000 s of particles per cm^3)	PM2.5 ($\mu g\,m^{-3}$)	NO (ppb)	Black carbon ($\mu g\,m^{-3}$)	CO (ppm)	CO_2 (ppm)	n
Long beach residential	26 (25, 28)	17 (15, 20)	17 (14, 21)	1.5 (1.1, 1.6)	0.13 (0.10, 0.18)	400 (390, 420)	329
Pasadena residential	13 (12, 15)	7.9 (7.3, 8.8)	16 (14, 19)	0.74 (0.39, 0.89)	0.13 (0.10, 0.27)	560 (540, 580)	87
USC start	43 (33, 53)	45 (44, 46)	59 (50, 66)	3.3 (2.6, 4.3)	0.54 (0.50, 0.58)	540 (540, 560)	41
USC finish	32 (31, 34)	23 (22, 24)	35 (32, 37)	2.2 (2.0, 2.6)	2.6 (2.4, 3.9)	580 (540, 620)	84
Arterial roads north of USC	33 (24, 53)	23 (20, 26)	79 (45, 120)	1.5 (1.2, 1.8)	1.8 (1.1, 2.5)	710 (690, 750)	117
110N Freeway (~3500 TrPD)	47 (33, 69)	25 (21, 29)	170 (120, 240)	2.4 (1.5, 3.6)	2.3 (1.8, 2.9)	820 (760, 860)	376
10E Freeway (~10,000 TrPD)	130 (95, 200)	110 (60, 820)	280 (210, 350)	13 (11, 20)	2.7 (2.3, 4.7)	930 (900, 980)	111
710S Freeway (~25,000 TrPD)	190 (150, 240)	54 (44, 60)	390 (330, 470)	12 (10, 16)	1.9 (1.6, 2.3)	850 (790, 910)	500

TrPD = average number of diesel-powered trucks per day (CalTrans, 2004), n = 4-day total number of 10-s averages in data set.

r^2's were reported by Bukowiecki et al. (2002a) between CO and UFP concentrations.

3.4. Instrument agreement, time series plots, and peak concentrations

UFP concentrations were very high on freeways, often an order of magnitude or more above concentrations in residential areas. This is illustrated in Fig. 4, showing 10-s average UFP concentrations for a 90 min run. Fig. 4 shows the typically high variability in UFP concentrations on freeways, where individual vehicle exhaust plumes sometimes caused sharp peaks in concentrations. Peaks "a" through "g" resulted from directly encountering the exhaust of a vehicle being followed, as determined from video records, similar to that observed in other studies (Fruin et al., 2004; Pirjola et al., 2004; Weijers et al., 2004; Bukowiecki et al., 2002a). These sharp peaks often occurred behind diesel-powered vehicles, although peaks "a" and "g" were from gasoline-powered vehicles, and the origin of many peaks were not readily identifiable. The sudden concentration changes between the freeways and the residential areas reflect the steep UFP concentration gradients found near freeways (e.g., Zhu et al., 2002a, b).

Figs. 5–7 demonstrate the excellent agreement between related pollutants, in this case presented in 60-s averages to improve the clarity of the graphs. The 3007 CPC UFP number concentrations and the EAD particle length measurements showed nearly perfect relative agreement, as shown in Fig. 5. Fig. 6 similarly shows excellent agreement between UFP and NO concentrations although NO concentrations relative to UFP become very low in residential areas due to the low numbers of diesel vehicles and the conversion of NO to NO_2. NO_2 showed much less variability, ranging from about 25 ppb in residential areas to about 100 ppb on the 710 Freeway, similar to measurements made by Seakins et al. (2002).

Another pollutant pair that showed excellent agreement was BC and PM-PAHs, as shown in Fig. 7. The data gap for BC from about 12:12–12:22 was due to a tape advance occurring in the Aethalometer, which occurred when the tape was saturated. It is interesting that for BC and PM-PAH, the time spent on the 110N Freeway, from 12:57 to 13:13, and on the 110S Freeway from 13:20 to 13:31, were almost indistinguishable from the time spent on Pasadena residential streets in between (13:13 and 13:20). This is a likely due to the similarly low diesel truck traffic on both the 110 Freeway and residential street in Pasadena.

In contrast to UFP and the pollutants strongly correlated pollutants (BC, NO, and PM-PAH), CO and CO_2 concentrations showed smaller differences between freeway segments and between freeways and arterial roads (Fig. 8). CO and CO_2 appeared to be most

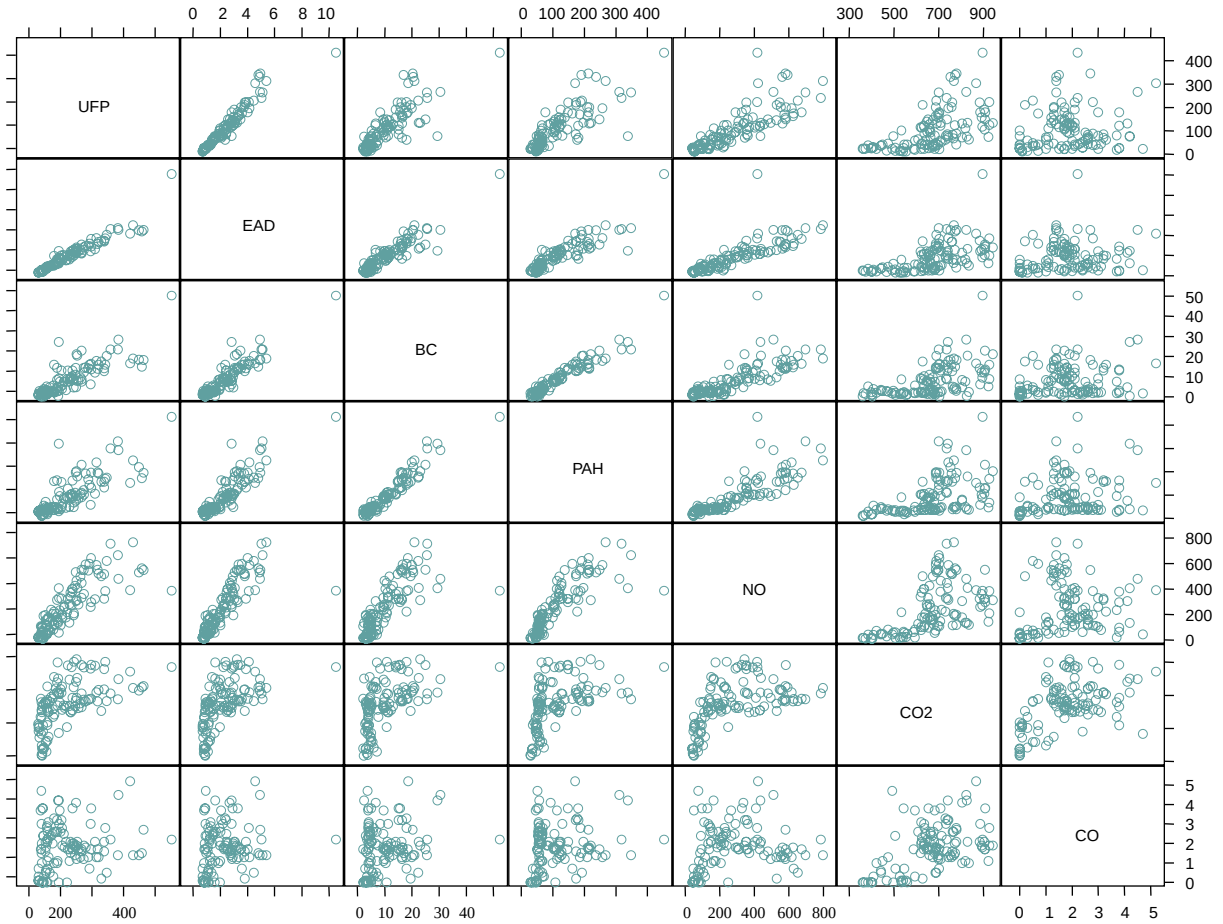

Fig. 3. Scatter plot matrix of 60-s average concentrations on 16 April. Units are 1000 s of particles cm^{-3} for UFP, mm cm^{-3} for the EAD, $\mu g\,m^{-3}$ for BC, $ng\,m^{-3}$ for PAH, ppb for NO, and ppm for CO_2 and CO. Note x and y axes are identical for each pollutant.

Table 4
The Spearman correlation ρ^2/Pearson's correlation r^2

	EAD	BC	NO	PM-PAH	CO_2	CO
UFP	0.98/0.88	0.88/0.76	0.90/0.78	0.88/0.69	0.68/0.53	0.20/0.21
EAD		0.92/0.75	0.93/0.69	0.93/0.71	0.69/0.47	0.25/0.20
BC			0.87/0.76	0.95/0.82	0.62/0.55	0.18/0.20
NO				0.93/0.73	0.65/0.55	0.28/0.23
PM-PAH					0.66/0.45	0.29/0.23
CO_2						0.46/0.49

affected by proximity to general traffic density. The instrument used for CO and CO_2 measurements also had a slower response time than other instruments, which reduced the apparent differences between locations or roadways if time on them was short.

3.5. Comparisons to other mobile platform studies

Table 5 shows a comparison of our results with other similar mobile platform studies. Our peak UFP concentrations of about 800,000 cm^{-3} generally exceeded

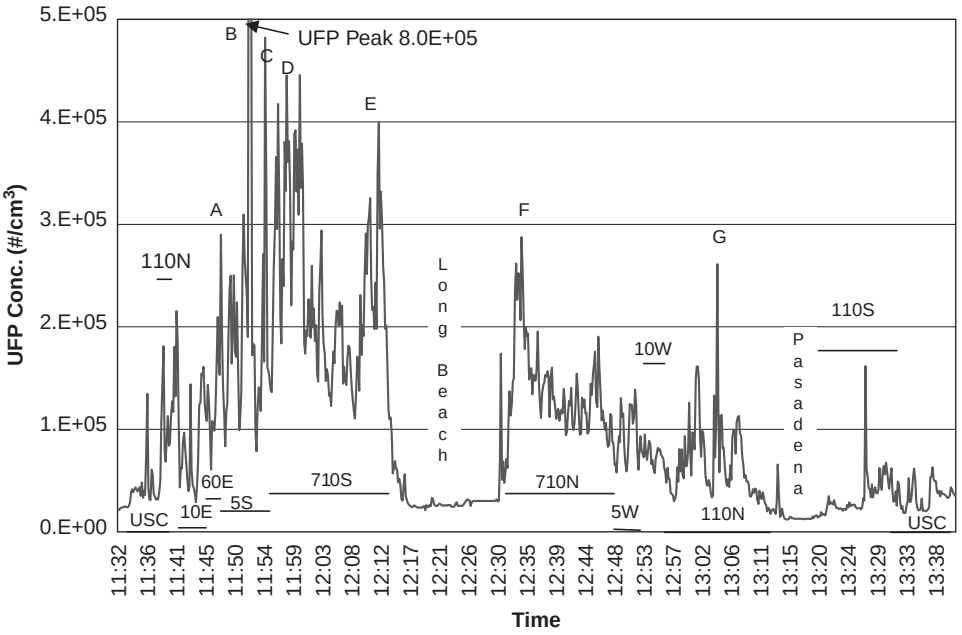

Fig. 4. UFP concentrations in various locations. Letter b through e correspond to directly following diesel-powered vehicles, while letters a and f correspond to impacts of gasoline-powered vehicles. Freeway segments are identified under data display.

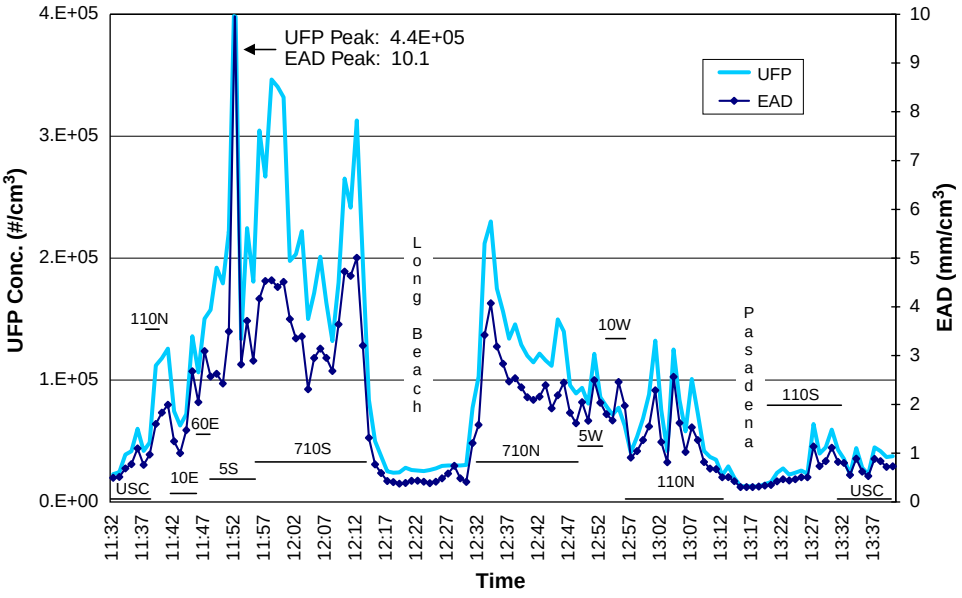

Fig. 5. Time-series plot of particle counts and particle length concentrations during April 16 freeway loop.

the results of other studies. These differences were likely due to our much higher traffic and diesel traffic volumes. The exceptions were tunnel measurements, or in the case of Kittelson et al. (2004a), where the CPC measured particles down to 3 nm rather than 7 nm, encompassing a size range with high numbers of particles.

The higher traffic volumes of our study are also reflected in our peak NO_2 and CO concentrations (the highest reported), and the highest reported roadway-average NO_x, and CO_2 concentrations. This is perhaps best illustrated by the CO_2 measurements—the 400 ppm roadway average reported in Kittelson et al. (2004a,b)

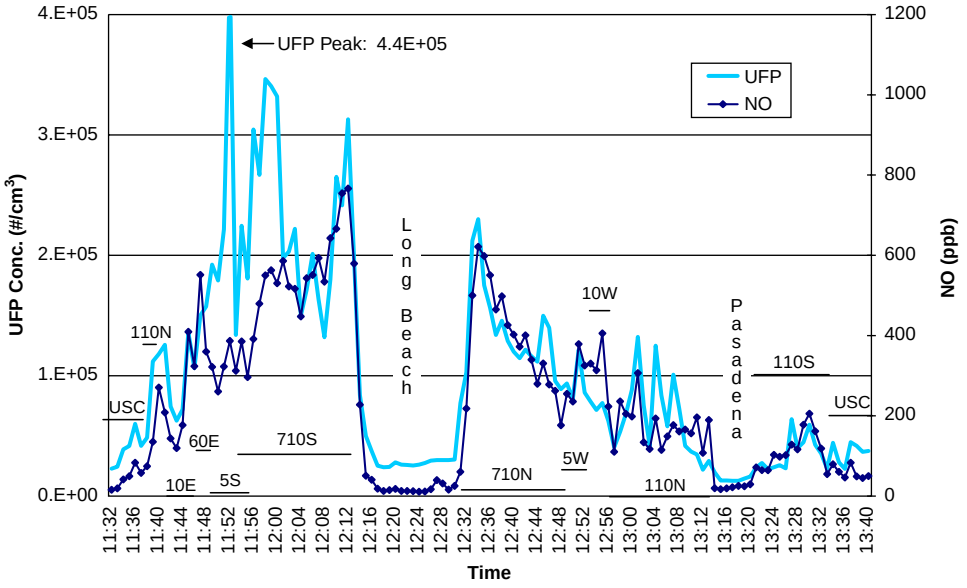

Fig. 6. Time series plot of UFP and NO concentrations during 16 April freeway loop.

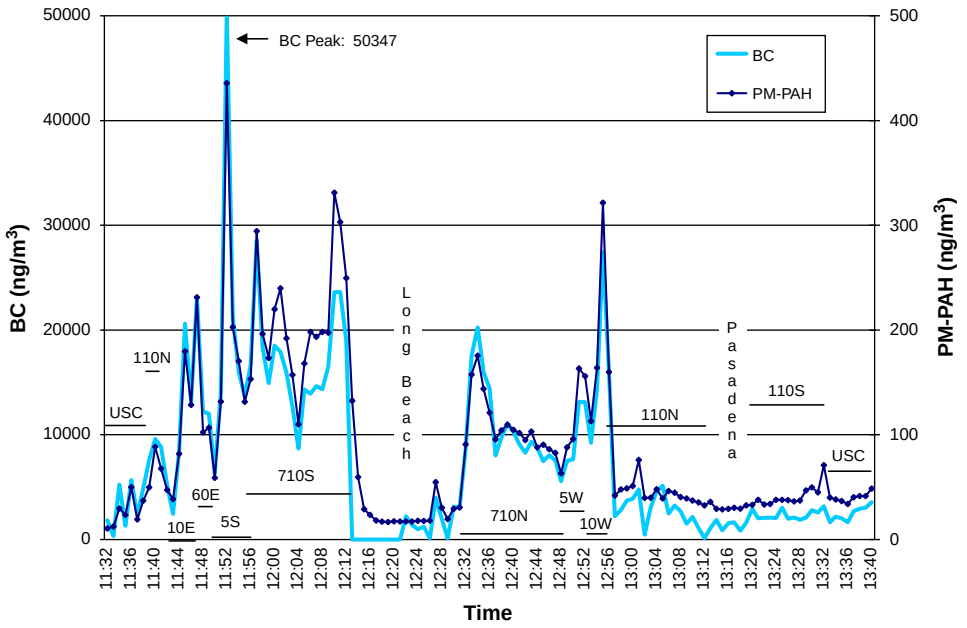

Fig. 7. Time-series plot of black carbon and PM-PAH concentrations during 16 April freeway loop.

was only 26 ppm above their measured background, while our CO_2 averages on roadways were several hundred ppm above our background (residential) concentrations. At the same time, our background concentrations for all comparable pollutants agree with those of the other studies.

3.6. UFP size distributions

Average UFP size distributions for the 110 Freeway (ten runs) and 710 Freeway (11 runs) on 16 April are shown in Fig. 9. These represent the freeway average of concurrent, 60-s scans made with two SMPSs. One

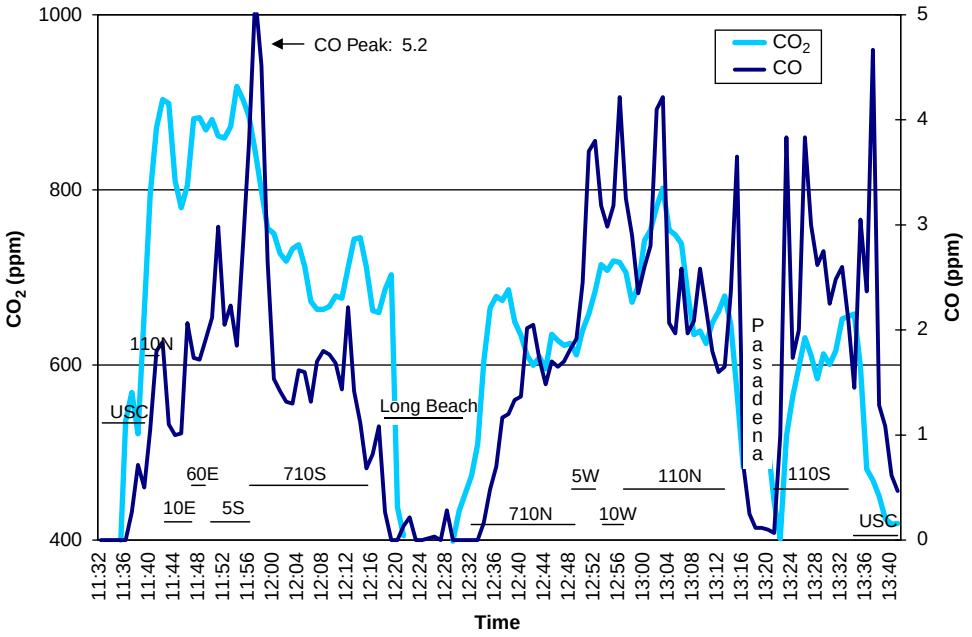

Fig. 8. Time-series plot for CO_2 and CO concentrations during 16 April freeway loop.

SMPS used a long DMA, Model 3081, which covered the particle size range from 0.017 to 570 μm, and the other SMPS used a nano-DMA, Model 3085, which covered particle sizes from 0.005 to 160 μm. The particle size range of 0.03–0.15 μm covered by both DMAs showed excellent agreement, differing by <5% on the 710% and 6% on the 110. Below 0.02 μm, however, the results diverged. It is likely that significant particle losses occurred in the long DMA below 0.03 μm. It is also apparent from this figure that when characterizing particle size distributions on roadways, the use of two SMPSs is advisable, although a single SMPS with a nano-DMA covers the UFP size range, i.e. particles < 0.10 μm.

The 710 Freeway, with 2–3 times greater UFP concentrations than other freeways, can also be seen to show a sharp UFP number mode near 0.02 μm, reflecting high traffic emissions, similar to results reported in Helsinki (Pirjola et al., 2004), Zurich (Bukowiecki et al., 2002a) Minneapolis (Kittelson et al., 2004a) and Los Angeles (Zhu et al., 2002b). The accumulation mode (0.1–1.0 μm) number concentrations are markedly less for the 110, as expected with much less diesel vehicle traffic (Kittelson, 1998; Kleeman et al., 2000). A future paper will be devoted to the nature of particle size and other physical parameters reported by the SMPS on freeways, arterial roads, and on residential streets.

3.7. Instrument reliability and performance

In general, instrument performance and reliability were found to be good in spite of the vibrations and accelerations sometimes present in a mobile environment. The 3022A CPC was more sensitive to vibration in this study than any other instrument used because it uses a reservoir of butanol as a source of super-saturated vapor to grow particles to optical detection range. Any significant tilt or acceleration can cause butanol to be pulled into the optics. This condition was avoided by filling and emptying the 3022A reservoir before each run to saturate the felt lining of the reservoir without keeping the reservoir full.

Other instruments were virtually trouble-free. No operational problems were encountered with the NO_x analyzer, the EAD, the Q-Trak, or the portable 3007 CPC. The Ecochem PM-PAH monitor required the replacement of its internal light source, and the Aethalometer was inoperable one day after post-rain humidity appeared to condense in the instrument optics after an overnight temperature drop. Disassembly and drying the next day solved this problem.

4. Conclusion

This study demonstrated the successful integration of sophisticated air pollution measurement instrumentation with a mobile platform. This creates a new tool for evaluating pollutants of concern in diverse and largely unstudied microenvironments. The mobile monitoring platform was tested in a study of Los Angeles freeways, arterial roads, and residential streets. In spite of the power and space limitations, and sometimes significant

Table 5
Comparison of reported peak and average concentrations for UFP and other gaseous pollutants for roadways and background locations

	This study	Kittelson et al. (2004a, b)	Bukowiecki et al. (2002a)	Weijers et al. (2004)	Pirjola et al. (2004)
City (locations)[a]	Los Angeles (U, C)[a]	Minneapolis (U, S, R)[a]	Zurich (U, S, R)[a]	Amsterdam (U, R, C)[a]	Helsinki (U, S)[a]
Roadway peaks:					
UFP (1000 s cm^{-3})	(to 7 nm)	(to 3 nm)	(to 3 nm)	(to 7 nm)	(to 3 nm)
Following diesels	800	2000	400	600	200–500
NO_2 (ppb)	200		60		
CO (ppm)	14		4		
Roadway avgs:					
UFP (1000 cm^{-3})	55–200 (F)[b]	400	30–50 (A)		
	40 (A)[c]	160			
NO_x (ppb)	230–470 (F)[b]				
	140 (A)[c]				
CO_2 (ppm)	800–900 (F)[b]	400			
	720 (A)[c]				
Background concs:					
UFP (1000 s cm^{-3})	14–27 Residential	9 Residential	5–15 (R)		<20–35
CO (ppm)	0.1–0.4[d]		0.2–0.4		10 night
CO_2 (ppm)	368–475[d]	364			
NO_x (ppb)	36–50[d]	15			

[a]*Note*: U = urban, S = suburban, R = rural/exurban, C = coastal; F = freeway; A = arterial.
[b]Range by freeway segment, 110N–710S, all 4 days.
[c]Arterial roads north of USC.
[d]Residential Long Beach location.

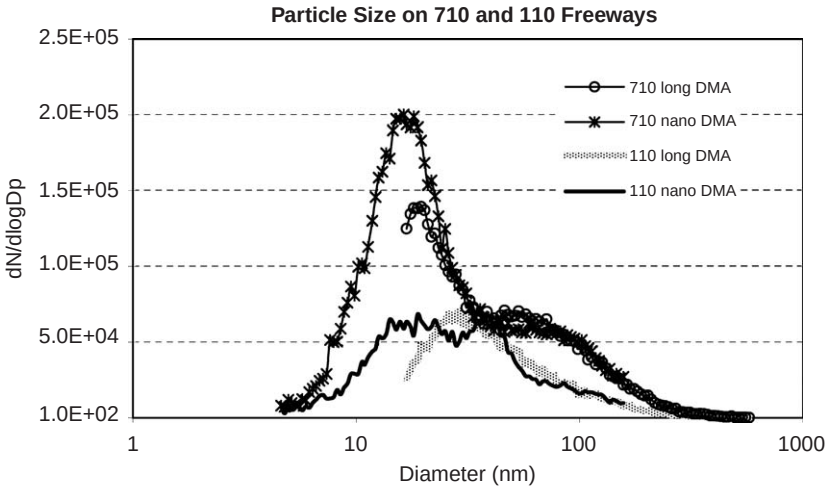

Fig. 9. Average particle size distribution on the 710 and 110 freeways, 24 April.

vibration, the instruments functioned reliably and showed excellent correlation between UFP count concentrations and BC, NO, and PM-PAHs. Freeway concentrations were frequently an order of magnitude higher than on residential streets for UFP, NO, BC, and CO. Future studies using mobile monitoring platforms are needed to fully evaluate pollutants in the micro-environments measured in this study. Studies should

focus on correlating on-road observations with pollutant concentrations, and exploring particle size and pollutant relationships. The findings of the current study will provide useful data to aid in the development of these efforts.

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